PB87-174991

Air Pollution Models as Descriptors of Cause-Effect Relationships

(U.S.) Environmental Protection Agency
Research Triangle Park, NC

Apr 83

U.S. Department of Commerce
National Technical Information Service
NTIS

TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completir

1. REPORT NO.	2.	3. PB87-174991
EPA/600/J-84/397		

4. TITLE AND SUBTITLE	5. REPORT DATE
AIR POLLUTION MODELS AS DESCRIPTORS OF CAUSE-EFFECT RELATIONSHIPS (Journal Version)	1984
	6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S)	8. PERFORMING ORGANIZATION REPORT NO.
Robert G. Lamb	
9. PERFORMING ORGANIZATION NAME AND ADDRESS	10. PROGRAM ELEMENT NO.
Same as block 12	CDWA1A/02-1335 (FY-84)
	11. CONTRACT/GRANT NO.
12. SPONSORING AGENCY NAME AND ADDRESS	13. TYPE OF REPORT AND PERIOD COVERED
Atmospheric Sciences Research Laboratory Office of Research and Development U.S. Environmental Protection Agency Research Triangle Park, NC 27711	14. SPONSORING AGENCY CODE EPA/600/09

15. SUPPLEMENTARY NOTES

Atmospheric Environment, 18(3):591-606, March 1984

16. ABSTRACT

The problem of air pollution modeling is treated beginning from a philosophical standpoint, in which a model is viewed as a "universal statement" and a complementary set of "singular statements" from which specific cause-effect relationships are deduced; proceeding to the formulation of a specific model from fundamental physical principles. In the course of the analyses, a number of basic issues are examined. These include the types of information that an air pollution model is capable of providing (it is shown that specific events are not predictable, only the set of possible events can be described); the problem of model "validation" (even with a perfect model and error-free input data and observations, discrepancies will exist between predicted and observed quantities); the character and representation of long-range dispersion (the conventional concepts of transport and diffusion become ill-defined when applied within the context of long-range dispersion models); and other topics relevant to the use of models in decision making processes.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group

REPRODUCED BY
U.S. DEPARTMENT OF COMMERCE
NATIONAL TECHNICAL
INFORMATION SERVICE
SPRINGFIELD, VA 22161

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS *(This Report)*	21. NO. OF PAGES
RELEASE TO PUBLIC	UNCLASSIFIED	17
	20. SECURITY CLASS *(This page)* UNCLASSIFIED	22. PRICE

EPA Form 2220-1 (9-73)

EPA/600/J-84/397
JOURNAL ARTICLE

Atmospheric Environment Vol. 18, No. 3, pp. 591–606, 1984
Printed in Great Britain.

0004-6981/84 $3.00 + 0.00
Pergamon Press Ltd.

AIR POLLUTION MODELS AS DESCRIPTORS OF CAUSE–EFFECT RELATIONSHIPS

ROBERT G. LAMB

Meteorology and Assessment Division, Environmental Sciences Research Laboratory, Research Triangle Park, NC 27711, U.S.A.

(*First received* 7 *December* 1982 *and received for publication* 21 *April* 1983)

Abstract—The problem of air pollution modeling is treated beginning from a philosophical standpoint, in which a model is viewed as a 'universal statement' and a complementary set of 'singular statements' from which specific cause–effect relationships are deduced; proceeding to the formulation of a specific model from fundamental physical principles. In the course of the analyses, a number of basic issues are examined. These include the types of information that an air pollution model is capable of providing (it is shown that specific events are not predictable, only the *set* of possible events can be described); the problem of model 'validation' (even with a perfect model and error-free input data and observations, discrepancies will exist between predicted and observed quantities); the character and representation of long-range dispersion (the conventional concepts of transport and diffusion become ill-defined when applied within the context of long-range dispersion models); and other topics relevant to the use of models in decision making processes.

1. INTRODUCTION

In environmental science, as in most areas of applied science, we are interested in cause and effect relationships, or in causal explanations of events. The desire for this knowledge stems from a need to know what actions (causes) we must take to achieve a particular change (effect) in environmental quality.

The philosopher Karl Popper (1959) noted that causal explanations require two ingredients: universal statements, which are hypotheses of the character of natural laws; and singular statements, which pertain to the specific event in question. It is from the *set* of these two types of statements that we deduce singular statements, or specific predictions, about events. The ability to make singular predictions is what is commonly referred to as 'determinism'.

In applied science we often use models as the basis of causal explanations. From Popper's viewpoint we see that a model is a universal statement. A theoretical model* is assembled from the fundamental physical laws of science and it generally takes the form of one or several differential equations. In this case the necessary complement of singular statements consists of the initial and boundary conditions on the equations and the values of the independent parameters that enter the equations.

It seems that a widely held view in many fields of modeling is that physical laws effect the transformation of some phenomena such as large scale weather systems, air pollutant transport, etc. into the realm of deterministic events, whereas certain other phenomena such as 'turbulence', dice games, etc. cannot be so transformed and retain a stochastic nature that is describable only in statistical terms. But from the points we have already raised it is clear that this perception of the role of physical law is not completely correct. Because in order to achieve determinism, i.e. in order to be able to make singular predictions, we need in addition to physical laws, i.e. universal statements, certain singular statements as well. This is apparent also in the fact that the mathematical equations that describe many natural phenomena are differential eqations that admit *infinitely* many solutions. It is only through the precise specification of certain initial and boundary conditions on the system that one effects the selection from the infinite set of possible solutions that single function that describes the system under the conditions prescribed. This is the essence of determinism.

It turns out, due to a combination of our inability to quantify the precise state of the atmosphere and its boundary and to the inherent instability of atmospheric motion, that not even large scale meteorological phenomena can be rendered deterministic. This fact has important implications for air pollution models and it is one of the main points that we will discuss in this paper.

Our principal objective is to illustrate how models, or universal statements, of air pollution phenomena can be assembled from the basic physical laws of science. However, once a model is developed, its meaningful use as a tool in decision making processes requires both an understanding of the inherent limits of model capabilities and some measure of the uncertainty associated with a particular model's predictions. For this reason we devote a considerable part of this paper to an analysis of model capabilities and uncertainties. This analysis begins in the next section where we consider some of the meteorological pro-

* We distinguish between theoretical and empirical models. The latter are inferred from a specific set of data or observations and generally they are not universally valid.

cesses that must be considered in the formulation of an air pollution model. As we noted earlier, it is in the treatment of meteorological phenomena that we encounter an inherent inability to make the singular statements needed to achieve determinism. This analysis continues into Section 3 where we examine the specific question of what aspects of air pollutant concentrations are predictable and what is required to predict them.

In Section 4 we describe how approximate descriptions of basic meteorological and chemical processes can be combined to formulate models for use in applied studies. The paper closes with a discussion of how a model should be used, the level of uncertainty in its predictions, what model 'validation' should entail, and associated matters related to the use of air pollution models as tools in air quality management.

In keeping with the spirit of this conference, the emphasis in this paper is on basic concepts and important issues rather than procedural details or mathematical techniques.

2. REPRESENTATION OF ATMOSPHERIC MOTION

It is obvious that atmospheric motion plays a major role in air pollution phenomena. In this section we consider the way in which we formulate a universal mathematical statement about the character of this role, and how this statement in conjunction with meteorological observations ultimately affect both the form of the diffusion model and the types of predictions that it can make.

In order to illustrate some basis concepts, it is helpful to consider first an elementary example. Consider the case of a single free particle in a gravitational field. By virtue of the law of gravitation and Newton's second law of motion, the altitude z of the particle must satisfy

$$\frac{d^2z}{dt^2} = g \tag{1}$$

where g is a known constant and t is time. The exact solution of (1) is

$$z(t) = \tfrac{1}{2}gt^2 + \dot{z}_0 t + z_0. \tag{2}$$

This expression defines an infinite set of functions $z(t)$ each of which has a different value of the constant z_0, which is the particle's position at time $t = 0$, and the constant $\dot{z}_0$, which is the particle's speed at $t = 0$. In other words, the physical laws embodied in (1) have not rendered the problem deterministic. They have delineated only a *global family* of possible particle trajectories $z(t)$ in space-time, as defined by (2).

Suppose that we have limited information about the particle, say we know that its initial position z_0 was $\hat{z}_0$, but we do not know its initial speed $\dot{z}_0$. The trajectory $z(t)$ is still not deterministic (we can do no better than guess at the position z at a given t) but we have reduced the set of possible trajectories from the global family (2) to the subfamily

$$z(t) = \tfrac{1}{2}gt^2 + \dot{z}_0 t + \hat{z}_0 \tag{3}$$

which we will call the *scenario family*. This subfamily, in which only $\dot{z}_0$ varies from member to member, is illustrated in Fig. 1 for the case $g = -19.6$ (relative units).

In this simple problem we have a universal statement, (1), which in conjunction with three singular statements, namely the value of g, the initial particle position z_0, and the initial particle speed $\dot{z}_0$, allows us to make singular (deterministic) predictions about the particle's position. But if we are unable to make all three of the required singular statements, we cannot deduce a singular statement about the particle's position. We can deduce only that the position is one of a *set* of values whose bounds we can specify.

In order to establish a parallel between the example just described and the atmosphere, it is helpful to regard the single particle as comprising a 'system' whose state at any time t is specified by the variable $z(t)$. In general, the state of a system can be represented by a single point in a *phase space*. The phase space of the single particle system is 1-dimensional, the z axis. Under the constraint imposed on the system by the physical laws represented by (1), if the initial state of the system is $z_0 = \hat{z}_0$, then the phase space point that describes the system state must follow the curves defined by (3) and illustrated in Fig. 1.

The state of a 2-D atmosphere, which for simplicity we shall assume is dry and insentropic, is specified by the vector function

$$\mathbf{u}(\mathbf{x}, t) = [u(x, y, t), v(x, y, t)] \tag{4}$$

where u and v are the x and y components of the fluid speed at time t at point $\mathbf{x}$. The phase space of this atmosphere is infinitely dimensional. It contains a coordinate axis for u and one for v for *each* of the infinitely many fixed points $\mathbf{x}$ that lie within the fluid.

The permissible states of this atmosphere are limited by two physical laws: conservation of mass, which can be expressed in the form

$$\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} = 0 \tag{5}$$

and the second law of motion, which can be expressed as

$$\frac{\partial \zeta}{\partial t} + u\frac{\partial \zeta}{\partial x} + v\frac{\partial \zeta}{\partial y} + v\frac{\partial f}{\partial y} = \kappa\left[\frac{\partial^2 \zeta}{\partial x^2} + \frac{\partial^2 \zeta}{\partial y^2}\right] \tag{6a}$$

where ζ is the vorticity, defined by

$$\zeta \equiv \frac{\partial v}{\partial x} - \frac{\partial u}{\partial y}, \tag{6b}$$

and f and κ are the Coriolis parameter and molecular viscosity, respectively, both of which are given, independent parameters. (Meteorologists will recognize (6a), when $\kappa = 0$, as the barotropic vorticity equation.) Equations (5) and (6) have infinitely many solutions

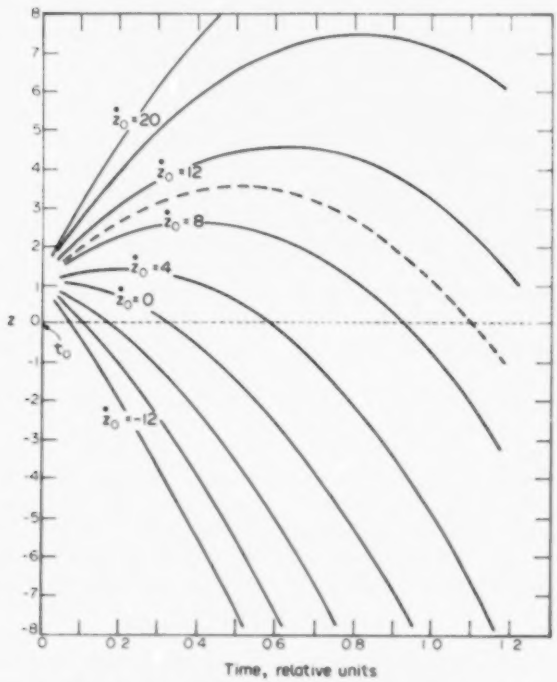

Fig. 1. Selected members of the scenario family of possible trajectories $z(t)$ of a particle located at time 0 at $z(0) = \hat{z}_0 = 1$ and whose position z satisfies Equation (1).

$\mathbf{u}(\mathbf{x}, t)$. As before we shall refer to this set of solutions as the *global family* of possible system states.

We say that a system is deterministic, rather than stochastic, if with error-free observations we are able to predict its state exactly at a given time t. We found that the single particle system was deterministic only if the initial position and speed of the particle (or equivalent information) were known. With less information the system becomes stochastic because we can specify its state only to within an infinite subset of functions in the global family of possible states.

In our 2-D atmosphere determinism requires the specification of the complete velocity field $\mathbf{u}(\mathbf{x}, t_0)$ at some instant t_0. Our ability to quantify the state of the atmosphere is limited to discrete measurements at a finite set of locations. Suppose that we have *error-free* measurements of the 2-D fluid speed $(\hat{u}_n, \hat{v}_n)$ at N arbitrarily located sites $\mathbf{x}_n$, $n = 1, \ldots N$ at some instant t_0. These data are not adequate to specify $\mathbf{u}(\mathbf{x}, t_0)$ completely. Rather we can say only that

$$\mathbf{u}(\mathbf{x}_n, t_0) = (\hat{u}_n, \hat{v}_n) \quad n = 1, 2, \ldots N, \tag{7}$$

where $(\hat{u}_n, \hat{v}_n)$ are given numbers. In addition to this information we also know that in accordance with (5). $\mathbf{u}(\mathbf{x}, t_0)$ is nondivergent, i.e.

$$\frac{\partial}{\partial x} u(\mathbf{x}, t_0) + \frac{\partial}{\partial y} v(\mathbf{x}, t_0) = 0. \tag{8}$$

Equations (7) and (8) define an infinite set of functions *one* member of which describes the actual flow field $\mathbf{u}(\mathbf{x}, t_0)$ that existed at the instant t_0 that the observations $(\hat{u}_n, \hat{v}_n)$ contained in (7) were made. In terms of our earlier terminology, the function set that satisfies both (7) and (8) is the *scenario family* of possible initial states. No matter how large N might be, as long as it is finite the set of equations (7) and (8) are not sufficient to describe the initial state of the flow field uniquely. Instead these equations define a set of functions any one of which could describe the actual state under the conditions observed. We wish to emphasize that the existence of more than one possible state is *not* due to errors in the observations or to inaccuracies in solving the governing differential equations. It arises from the fact that the system has more degrees of freedom than we are able to specify. The single particle system discussed earlier has two degrees of freedom: z_0 and $\dot{z}_0$. Specifying only the former left open the possibility of the infinitely many states illustrated in Fig. 1, and defined by Equation (3). The atmosphere has infinitely many degrees of freedom and hence we conclude that *atmospheric motion is non-deterministic*. Later we will show that within considerable limitations certain space and/or time averaged properties of the state of the atmosphere, and also of material dispersion, can be predicted approximately.

We will denote the subset of functions $\mathbf{u}(\mathbf{x}, t_0)$ that

satisfy (7) and (8), i.e. the scenario family, by

$$W^N(t_0) \equiv \text{set of } \mathbf{u}(\mathbf{x}, t_0),\ \mathbf{x} \in D, \text{ that satisfy (7) and (8);} \tag{9}$$

where D represents the entire spatial domain occupied by the fluid. (For simplicity we will assume that D is the surface of a sphere so that we do not have to be concerned with lateral boundary conditions.) The superscript N on W signifies that the definition of the function set is conditioned on N observations, as indicated explicitly in (7).

The set $W^N(t_0)$ defines a hypersurface in the phase space of atmospheric states. Each point on this surface represents an entire vector function $\mathbf{u}(\mathbf{x}, t)$, $\mathbf{x} \in D$, $t = t_0$.

The solutions of the governing equations (5) and (6) define curves in phase space which we will call *state trajectories*. Since the solutions of (5) and (6) with $\mathbf{u}(\mathbf{x}, t_0)$ given are assumed to be unique, one and only one state trajectory passes through each point in $W^N(t_0)$. Thus, by following each of the state trajectories forward in time from its origin in $W^N(t_0)$ to a time $t = t_0 + \Delta t$, we define a new function set $W^N_{\Delta t}(t_0)$:

$$W^N_{\Delta t}(t_0) \equiv \text{set of end points of state trajectories of Equations (5) and (6) of time span } \Delta t \text{ originating in } W^N(t_0). \tag{10}$$

This set represents the set of possible atmospheric states at time $t_0 + \Delta t$. The sets W^N and $W^N_{\Delta t}$ and the state trajectories that connect them are illustrated schematically in Fig. 2.

Despite the fact that the set of observations (7) and the continuity equation (8) do not define a unique fluid state $\mathbf{u}(\mathbf{x}, t_0)$, the common quest in meteorological modeling is for a so-called objective analysis scheme that can extract from these equations a single 'best' estimate of the actual state $\mathbf{u}(\mathbf{x}, t_0)$ that existed at the moment the observations were taken. Deriving a single function from (7) and (8) requires the imposition of additional constraints adequate to transform this underdetermined set of equations into one possessing a single solution. In short, a closure approximation is required. One of the simplest closure approximations used in objective analysis procedures is to assume that the velocity $\mathbf{u}(\mathbf{x}, t)$ at each point where observations were *not* made is some specified weighted averaged of the observed values at the closest surrounding measurement sites. But this assumption introduces artificial information into the problem inasmuch as it is not based on investigations of the specific atmospheric problem under study. The fact remains that the only real way to improve our ability to specify the state of a system is by increasing our number of observations of it. We return to this point later.

Regardless of how many observations we acquire, we can never specify the state of a system with infinitely many degrees of freedom, like the atmosphere, better than to within a set of possible states. It turns out that this limitation places significant restrictions on our ability to make long-range predictions of the state of the system. Because Lorenz and others (see Dold and Eckmann, 1977) have shown mathematically that nonlinear systems characterized by equations of the form of (6) are so inherently unstable for certain parameter values (namely large Reynolds numbers in the case of fluids), the state trajectories that pass through two arbitrarily close initial state $\mathbf{u}(\mathbf{x}, t_0)_1$ and $\mathbf{u}(\mathbf{x}, t_0)_2$ diverge in phase space so rapidly (see Fig. 2) that after some finite time Δt the states $\mathbf{u}(\mathbf{x}, t_0 + \Delta t)_1$ and $\mathbf{u}(\mathbf{x}, t_0 + \Delta t)_2$ bear almost no resemblence to each other. This means that even if we were working with numerous, precise observations and exact solutions of the governing equations, our inability to specify $\mathbf{u}(\mathbf{x}, t_0)$ in its totality would result eventually in a complete failure of the model forecast.

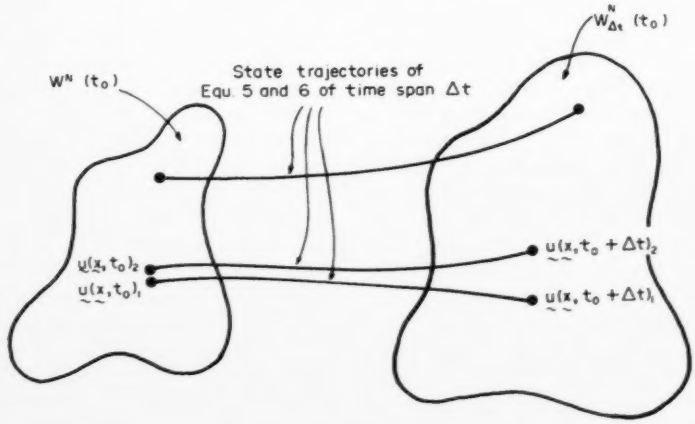

Fig. 2. Illustration of the function sets $W^N(t_0)$, defined by (9), and its forward projection $W^N_{\Delta t}(t_0)$ defined by (10), both in the phase space of the 2-D fluid states $\mathbf{u}(\mathbf{x}, t)$. In this space the solutions of Equations (5) and (6), which represent the physical laws obeyed by the fluid, are curves which we call 'state trajectories'.

All of the points raised above emphasize that in applied studies it is meaningless to discuss the state of the atmosphere in terms of single functions. All we can hope to do is to narrow the set of functions that constitutes all possible states. And as we noted earlier, this task requires additional observations.

Additional observations are generally readily available from the N fixed sites at times following t_0. This is especially true in dispersion studies where we are most often interested in meteorological episodes, or scenarios, in which we have observations of the atmosphere not only at one instant t_0, but also at subsequent times $t_1 (> t_0), t_2, \ldots t_M$ during the period $t_0 \leqslant t \leqslant t_0 + \mathcal{T}$ of concern. Using a technique known as *pull back*, we can project the observations at times $t > t_0$ back along the state trajectories to the initial instant t_0 and effect a reduction in the size of the set of possible initial states. To see this in simple terms, consider again the earlier single particle problem. Suppose now that in addition to the initial observation of the particle's position $z(t_0) = \hat{z}_0 = 1$, we observe that at the later time $t_1 = 0.5$ the position is $z(t_1) = 3.5$. Since one and only one state trajectory passes through the phase space point (0.5, 3.5), namely the dashed line shown in Fig. 1, we can trace this line back to $t = t_0 = 0$ where we find that the initial particle speed must have been $\dot{z}_0 = 9.9$. In other words, subsequent observations of the state of the system tell us which of the state trajectories the actual system is *not* following. By tracing those trajectories back to the initial moment t_0, we delineate a subset of states that we can rule out as possible initial states. In the single particle problem which has only two degrees of freedom (for given g), the two observations of the system's state are sufficient to rule out all initial states but one and hence in this case the system is rendered deterministic.

Turning now to the case of the 2-D atmosphere, suppose that in addition to the N observations of velocity at t_0 we have N observations (not necessarily at the same sites) of velocity at time $t_0 + \Delta t$. The latter define a function set $W^N(t_0 + \Delta t)$ analogous to $W^N(t_0)$, which is defined by (9). Since we know that the actual state of the atmosphere at time $t_0 + \Delta t$ is a member of the set $W^N(t_0 + \Delta t)$ and that it is also a member of $W^N_{\Delta t}(t_0)$, which is the forward projection of $W^N(t_0)$ defined by (10), it follows that the actual state must be a member of the intersection of these sets which we will call

$$W^{2N}_{\Delta t}(t_0) \equiv W^N(t_0 + \Delta t) \cap W^N_{\Delta t}(t_0). \tag{11}$$

In keeping with our convention for naming function sets, the superscript $2N$ signifies that $2N$ observations are incorporated in the delineation of the set. All of the sets described above are illustrated in Fig. 3.

If we 'pull back' the intersection set $W^{2N}_{\Delta t}(t_0)$ along the state trajectories that pass through it to time t_0, we define a subset $W^{2N}(t_0)$ of $W^N(t_0)$ which we know contains the actual system state at time t_0. In other words the observations at time $t_0 + \Delta t$ allow us to rule out as possible initial states all members of $W^N(t_0)$ outside the subset $W^{2N}(t_0)$ (see Fig. 3).

If we have N observations of the fluid velocity at time $t_0 + 2\Delta t$, these define a set $W^N(t_0 + 2\Delta t)$, through (9); and the intersection of this set with the forward projection of $W^{2N}(t_0)$ to $(t_0 + 2\Delta t)$ defines another set, namely

$$W^{3N}_{2\Delta t}(t_0) = W^N(t_0 + 2\Delta t) \cap W^{2N}_{2\Delta t}(t_0), \tag{12}$$

which contains the actual system state at time $t_0 + 2\Delta t$. The 'pull back' of $W^{3N}_{2\Delta t}(t_0)$ to t_0 defines the subset $W^{3N}(t_0)$ of $W^{2N}(t_0)$ illustrated in Fig. 3, that contains the actual fluid state at time t_0.

In general, with N, error-free observations of the state of a system at each of M times $t_0, t_1, \ldots, t_{M-1}$, one can assert unequivocally that the state of the system at time t_0 is a member of the function set $W^{MN}(t_0)$ defined as above; and that the state of the system at any time $t \geqslant t_0$ is a point on one of the state trajectories that

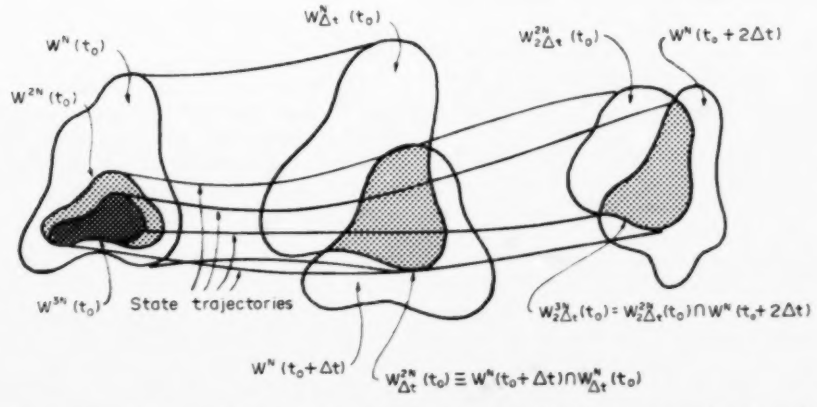

Fig. 3. Illustration in the phase space of the 2-D atmosphere of the function sets, state trajectories and pull back operations that describe the states of the atmosphere that are possible under the constraints of physical laws and the observations available.

emanate from the points in $W^{MN}(t_0)$. We call $W^{MN}(t_0)$ and its associated set of state trajectories the *scenario family* of possible atmospheric states.

In the context of diffusion modeling, the results obtained above indicate that even with perfect meteorological data and exact solutions of the equations of motion, we could specify the atmospheric flow field $\mathbf{u}(\mathbf{x}, t)$, $\mathbf{x} \in D$, $t > t_0$ only to within the set of functions represented in phase space by the state trajectories that pass through the points of the function set $W^{MN}(t_0)$ defined by physical laws and the MN observations available. Moreover, our analysis has not revealed any natural division of atmospheric motion into what one might regard as a stochastic part and a deterministic part. Indeed, we have shown that strictly speaking, *all* scales of atmospheric motion are non-deterministic. This result contradicts the conventional diffusion modeling practice of representing atmospheric motion as the composite of a 'mean', or deterministic, component of the flow $\bar{\mathbf{u}}$ and a stochastic 'turbulent' part $\mathbf{u}'$, i.e.

$$\mathbf{u}(\mathbf{x}, t) = \bar{\mathbf{u}}(\mathbf{x}, t) + \mathbf{u}'(\mathbf{x}, t) \tag{13}$$

where $\bar{\mathbf{u}}$ is simply an *ad hoc* fit of a continuous function to the set of MN wind observations. A scenario family mean exists, say $\langle \mathbf{u}(\mathbf{x}, t) \rangle$, but clearly it cannot be derived from the observations alone.

We propose, therefore, that in regional modeling the current concepts of 'turbulence' and 'mean flow' should be replaced by that of a single, non-deterministic class of flow. For the purpose of discussion in this paper, we will call the proposed fluid motion *metulence* and distinguish between different 'orders' of metulence. We define

$$\text{order} - MN \text{ metulence} \equiv \text{the set of flow fields } \mathbf{u}(\mathbf{x}, t), \tag{14}$$

$x \in D$, $t \geqslant t_0$ represented in phase space by the state trajectories of the equations of fluid motion that pass through the points of the function set $W^{MN}(t_0)$ whose members satisfy jointly physical laws and error-free observations of the fluid velocity at N arbitrary points in space at each of M different instants in time.

In Lamb (1983) it is shown mathematically that 'turbulence' as it is currently defined in the context of short-range, point source dispersion studies is a subset of order-1 metulence.

Consider now the general diffusion modeling problem where we wish to compute the concentrations of J chemically reactive pollutant species c_j, $j = 1, \ldots, J$ that will result from a *given* distribution $S_j(\mathbf{x}, t)$ of sources of these species under a particular meteorological situation described by N observations of the flow at M different times in a spatial domain $\mathscr{D}$ and time interval $t_0 \leqslant t \leqslant t_0 + T$. In an analogy with the fluid phase space, we define an infinite-dimensional *concentration phase space* which contains one coordinate axis for each of the J species concentrations at each of the infinitely many points $\mathbf{x}$ in $\mathscr{D}$. A point in concentration phase space represents a vector function $[c_1(\mathbf{x}, t), \ldots, c_J(\mathbf{x}, t)]$, $\mathbf{x} \in \mathscr{D}$, t = given instant. The evolution of the concentration distributions in time is represented by trajectories in concentration phase space that are defined by the solution of the system of differential equations

$$\frac{\partial c_m}{\partial t} + \nabla \cdot \mathbf{u} c_m \equiv \kappa_m \nabla^2 c_m + \sum_{i=1}^{J} \sum_{j=1}^{J} k_{mij} c_i c_j + S_m, \quad m = 1, \ldots, J \tag{15}$$

which express both the physical law of mass conservation and the chemical law of species transformations. In (15), κ_m denotes the *molecular* diffusivity of species m, k_{mij} are elements of the chemical rate coefficient matrix and $\mathbf{u}(\mathbf{x}, t)$ is the fluid velocity which, in the present instance, is any one of the members of the set of order-MN metulence delineated by the available meteorological data. Thus, starting from the initial state

$$c_j(\mathbf{x}, t_0) = 0, \quad \mathbf{x} \in \mathscr{D}, \quad m = 1, 2, \ldots, J, \tag{16}$$

and with κ_j, k_{mij} and S_j *given*, (15) 'maps' each state trajectory in the order-MN metulence set in velocity phase space onto a unique state trajectory in concentration phase space. Stated another way, (15) maps each point in the scenario family of atmospheric states onto a point in a scenario family of concentration states. By virtue of (16) each of the state trajectories in concentration phase space emanates from the origin of the coordinate axes. The relationships between the scenario families in the velocity and concentration phase spaces are illustrated in Fig. 4.

In summary, we have shown that given precise meteorological data, exact solutions to the equations of motion, perfect emissions data S_j, total and precise knowledge of the species chemistry k_{mij} and exact solutions of the concentration equation (15), it is still impossible to predict the concentration levels that will result at any point or time with a resolution better than a *set* of values. Consequently, the utility of models in applied studies hinges on whether this set of possible values in a given problem is narrower than the error tolerance that one is willing to allow for the predictions. In the next section we consider this question. We have also indicated that in regional scale studies the phenomena of 'turbulence' and 'mean' motion are not well defined and we have proposed that both should be replaced by a single type of flow that for now we have called *metulence*. We gave a rigorous definition of metulence and pointed out that 'turbulence' as it is usually defined in short-range diffusion studies is a subset of order-1 metulence.

3. WHAT CAN MODELS PREDICT?

In practice models are based on approximate descriptions, or parameterizations, of physical laws; they employ approximate solutions of the governing equations; and they utilize values for the independent parameters that are derived from imperfect data. The

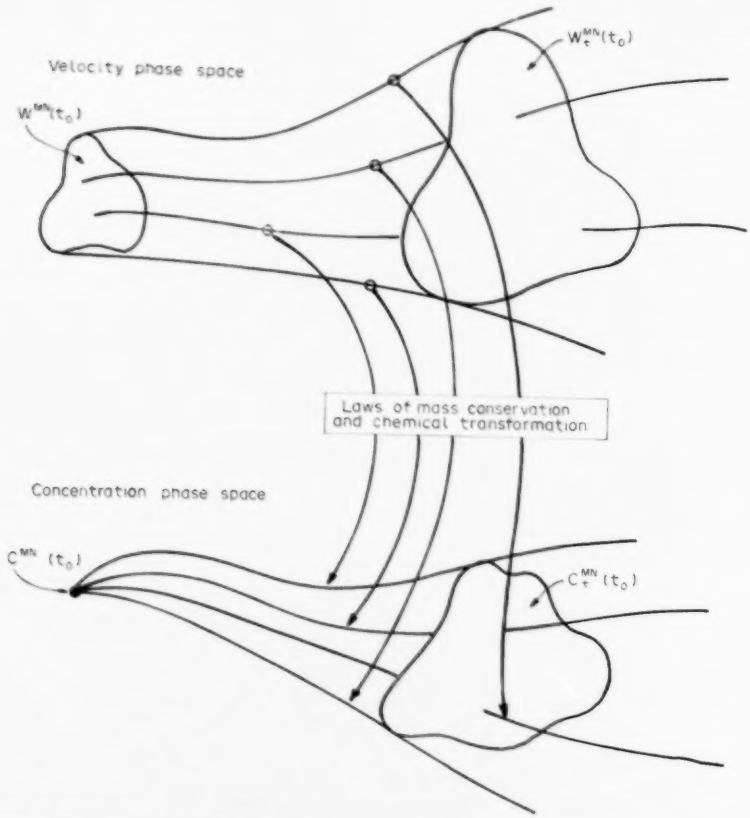

Fig. 4. Illustration of the link between the members of the scenario family of atmospheric states, $W^{MN}(t_0)$ and its state trajectories and the members of the scenario family of concentration states, $C^{MN}(t_0)$ and its state trajectories.

goal of model development is to reduce the degree of approximation in models and to refine the accuracy of the input data so that model predictions can be made more accurate. But suppose we managed to produce the perfect model, that is, one in which physical laws were described precisely, the input data were all error-free, and we had exact solutions of the governing equations. Would the uncertainties in the state of the atmosphere described in the previous section preclude our predicting a given quantity with the accuracy required? If so, then our effort to improve models is as futile as an attempt to develop a perpetual motion machine. In this section we explore the question of what a 'perfect' model could predict given the inherent ambiguity in the specification of atmospheric motion discussed above.

A game analogy of diffusion modeling

In order to help convey some basic ideas, we consider a simple game analogy that possesses many features in common with the diffusion modeling problem but which is less abstract. Our game involves the throwing of three dice three times to produce a 3×3 matrix of numbers **A**, say. (We consider 3 throws of 3 dice, rather than a single throw of 9 dice, as a way of retaining space-time features.) We assume for simplicity that the faces of each of the three dice are marked with an integer from the set 1, 2, 3, 4, 5 and 6. In this case the matrix **A** is a member of the family of $6^9 = 10{,}077{,}696$ matrices illustrated in Fig. 5. This family of matrices represents the scenario family of states of the dice 'system'. Like the scenario families of the velocity, i.e. $W^{MN}(t_0)$ and all points on its state trajectories, and the concentration systems, the matrices illustrated in Fig. 5 represent the set of all states of the dice system that are consistent with the information available, namely 3 dice; with faces marked 1, 2, 3, 4, 5 or 6; thrown 3 times.

A property common to all three systems is that none is deterministic. That is, we can delineate a set of states accessible to the system but we cannot specify exactly which of its states the system is in at any time. As we noted earlier, with our predictive capabilities limited to delineating sets of possible values, modeling will have little value in applied studies unless the range of values of quantities of interest within the set is narrower than

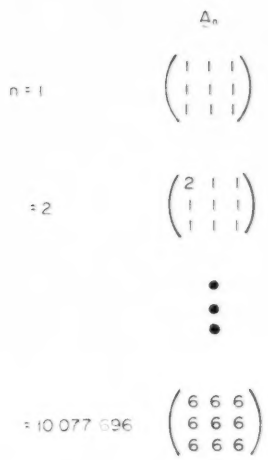

Fig. 5. The scenario family of outcome matrices **A** resulting from the throw of three dice three times (see text for more details).

the error level we are willing to tolerate in the prediction. For example, if we are interested in hourly averaged concentration at fixed points but the hourly averaged values of the members of the scenario family of concentration span a wide range of values, say a range as wide as the set of values we could *guess* might occur, than in such a case we would consider modeling to be a pointless exercise. Thus, it is important to determine which space-time averages, if any, of the members of the scenario sets of accessible states lie within a range of values narrower than the level of uncertainty allowable in the predictions. We explore this question first within the context of the dice system in order to illustrate basic concepts.

Let's assume that each column in **A** represents the numbers on each of the three dice on one of the three roles and is roughly analogous to three spatial points at a fixed time. Similarly, each row of **A** contains the 3 outcomes of each of the 3 successive roles and we will regard it as analogous to temporal variation at a fixed space point. To make the analogy between the dice and concentration states clearer, note that each matrix **A** would represent one state trajectory in a hypothetical problem involving a single species whose concentration could have only the integer values 1, 2, 3, 4, 5 or 6; space were 1-dimensional and quantized into 3 points; and the time period of interest consisted of only 3 distinct instants. The only flaw in this analogy is that the elements of the **A** matrix are independent parameters, whereas within a concentration field a certain functional relationship must exist, by virtue of the physical laws described by (15), between the concentration value at any point and the values within its immediate vicinity in space-time.

Within this frame of view the attribute "the sum of the 9 elements of **A**" is the analogue of a space-time average of one possible concentration distribution. Suppose we examine the family of $\mathbf{A}_n$ shown in Fig. 5 and compute the sum S_n of the 9 elements of each member, namely,

$$S_n = \sum_{i=1}^{3} \sum_{j=1}^{3} a_{ijn} \tag{17}$$

where a_{ijn} are the elements of $\mathbf{A}_n$.

We find that the sums S_n lie in the range $9 \leqslant S_n \leqslant 54$ for all n. Only one of the $\mathbf{A}_n$ has the sum 9, namely the member $n = 1$; and only the last member has the sum 54. Nine members have the sum 10 and nine possess the sum 53. But an inordinate 767,394 have the sum 31 and the same number have the sum 32. In fact, over half of the entire family have sums that are within $\pm 10\%$ of the family mean sum 31.5; and almost 90% have sums within $\pm 20\%$ of the family mean. The full distribution of sums S_n is plotted in Fig. 6. (We add in passing that in the 9-D phase space of the dice system, the set of points that represent the $\mathbf{A}_n$ whose element sums S_n have a given value comprise a hyperplane. The ordinate of Fig. 6 is a measure of the surface area of the hyperplane, associated with the S values on the abscissa, contained within the volume of phase space in which the system states are confined, namely $1 \leqslant a_{ij} \leqslant 6$, $i, j = 1, 2, 3$.)

The tight clustering of the sums S_n about the family mean value suggests that even though we cannot predict which of the matrices $\mathbf{A}_n$ will occur on any occasion, we can predict with respectable accuracy what the sum of its elements will be, *provided* that there is nothing inherent in the system that causes preferential occurrence of those matrices whose sums are farthest from the family mean value. Thus, *a priori* we still cannot assess the extent of our predictive capabilities in a given problem. We need more information about the system to do that. Realizing that the specification of the set of possible states, i.e. the scenario families, embodies all the information that is available about the system itself, we turn attention to the 'mechanism' that causes the system to acquire *particular* states.

In this endeavor we must keep in mind that up to now we have been discussing general systems. For example, the dice system that we have defined encompasses many different *specific* systems. The one that usually comes to mind is well balanced ('fair'), 6-sided dice with each face given a different number from the set of the first 6 integers. But under the same general definition we could also have dice with more than 6 faces, dice with the same number appearing on more than one of its faces, bizarre methods of throwing the dice, and so on. Similarly, the atmosphere that we are considering might be bounded below by terrain features or large bodies of water that bias flow directions in a particular way. In most cases the necessary information about the specific system is either unknown, e.g. we might not be allowed to see the dice that are thrown; or impossible to quantify, as is the

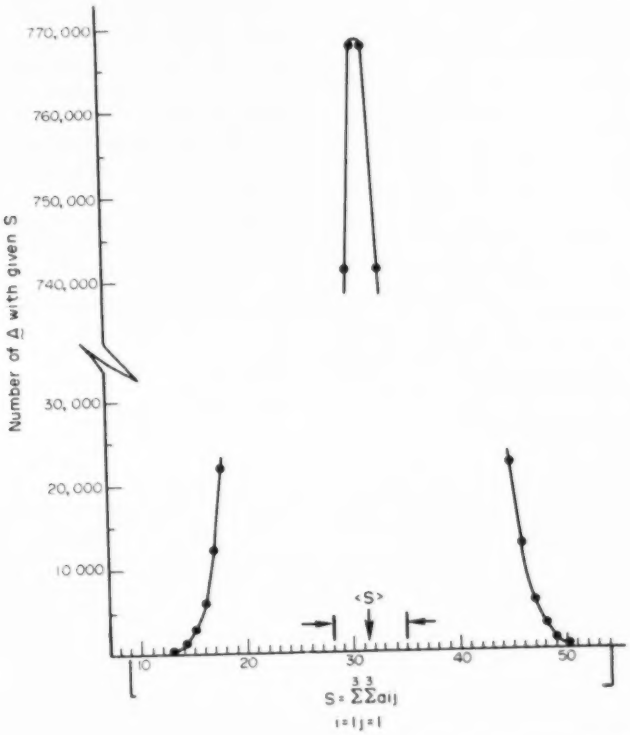

Fig. 6. Number of matrices in the scenario family with given element sum S.

case with the earth's surface texture or with the dynamical details of how the dice are held and thrown. In all such cases, we must treat the 'mechanism' that controls the system in a manner that accounts explicitly for the uncertainty that is associated with it.

Toward this end we imagine that in every system the controlling mechanism can be considered to be what we will call a *causal wheel* which we define as follows. For every *specific* system we imagine that there exists a well balanced wheel and pointer with numbers representing the members of the scenario family inscribed on the circumference of the wheel. Before each realization of the system, the wheel is spun (by mother nature) and the number indicated by the pointer when the wheel stops determines the state that the system acquires on that occasion. In this model our inability to predict the precise state of the system is simulated by the uncertainty in where the wheel will stop after it is spun, and biases and nonuniformities inherent in the *specific* system but unknown to us *a priori* are taken into account by the space allocated to each member of the scenario family on the causal wheel.

For example, using the above dice system to illustrate, we note that the most straightforward arrangement of the numbers n representing the members of the scenario family depicted in Fig. 5 is to include all the numbers, $n = 1, 2, \ldots, 10{,}077{,}696$ and to allocate equal angles of arc to each. This configuration is the one we would associate with 'fair' dice games in which any one of the possible outcomes (**A**) is as 'likely' to occur as any other. Another possible arrangement of the numbers is to include them all but to allocate more space to some numbers than others. This would represent a game in which the dice are 'loaded'. Yet another possibility is to exclude some numbers n altogether. If a specific set of dice were all of the common type in all respects except that one had a '5' on the face that normally would be marked with a '6', our corresponding 'causal wheel' would not contain the numbers n of Fig. 5 that denote matrices with 6's in row 2, say; and those numbers that represent matrices with 5's in row 2 would have twice the space allocations as the others.

In the context of the causal wheel, the *probability* of occurrence of a particular state n in the scenario family of possible states is defined simply by

$$\text{probability of state } n \equiv \frac{\lambda_n}{2\pi} \tag{18}$$

where λ_n is the arc length in radians allocated on the circumference of the wheel to member n of the scenario family. (We point out that there are a number of definitions of probability, see Papoulis (1965) and ours is very similar to what Papoulis refers to as the *axiomatic* definition.)

The concept of probability is an artifice for characterizing in a quantitative manner the collective effects of all the unknown or indescribable mechanisms or forces that control which of the possible states a system will acquire on a particular occasion. Thus, probabilities can only be inferred by watching the behavior of the specific system in question many times and keeping track of which states occur and how frequently they occur. A list of the states that are observed on each of many different occasions is called an *ensemble*. In terms of the 'causal wheel', an ensemble is generated by spinning the wheel many times and listing the n's that result. However, uncertainty seems to be self-perpetuating inasmuch as we find that not even the probabilities can be determined precisely, no matter how large an ensemble of observations we collect.

To see this, suppose we had a perfectly balanced wheel with the numbers $n = 1, 2, \ldots, N = 10{,}077{,}696$ arranged at equal intervals around its rim. Since there exists no force to cause the wheel to stop on each of the N numbers once and only once in N spins, an ensemble of N numbers produced by N spins might not contain some of the n. Obviously, we would be wrong if we inferred from this ensemble that the missing n were not present on the wheel. It is this same freedom of the action of the wheel that makes it impossible to predict the frequency with which given states of a system will occur.

For example, our intuition tells us that throwing nine dice and having them all come up six's, i.e. the last member of the **A** family of Fig. 5, is a 'rare' event. Yet if the dice are 'fair', this event is just as likely as any of the others. Thus, all of the outcomes are equally rare so a 'rare' event occurs on every outcome of the throw! It follows that there is no reason for 'rare' events to occur infrequently. In the language of hydrologists, the 'hundred year flood' may occur in several years in succession. Consequently, all we can do is base decisions on the *expected* frequencies of events, and then to hope that we're lucky!

The expected frequency of the all-6 matrix of Fig. 5 in a fair game is one in 10,077,696 games. If, in some hypothetical game, the player won \$10 on every throw that the all-6 matrix *did not* come up but was beheaded the first time it did, one could 'expect' to make a million dollars a year at this game for 100 years before suffering the ultimate loss. But there is nothing in the universe to prevent the all-6 matrix from coming up the very first game! Similarly, we can 'expect' the sum S of the ('fair') dice to differ from 31.5 by no more than $\pm 20\%$ in only one game in ten. But there is nothing to prevent exceedance in ten successive games. Our purpose in elaborating on these basic concepts is partly to emphasize the limits on the knowledge that we can hope to acquire about the future behavior of natural systems. We especially want to emphasize that the notion of determinism that underlies current thinking and practices in air pollution modeling and in the expectations that regulatory officials have in the utility of model predictions is unsupportable.

Having collected an ensemble of observed states of a system, we can compute *ensemble mean* values of given parameters by averaging the value of that parameter associated with each member of the ensemble. We will denote ensemble means by the angle brackets $\langle\ \rangle$. Intuitively, one can see that the ensemble mean can be computed from the scenario family by weighting the parameter value associated with each member of the family by the probability of that member, as defined in (18). For example, the ensemble mean of the matrix element sum S_n discussed earlier is

$$\langle S \rangle = \sum_{n=1}^{10{,}077{,}696} \text{Prob}\,(\mathbf{A}_n) S_n. \tag{19}$$

If S represents the sum of the elements of the matrix **A** that occurs on a given throw, then as we have already discussed, S is not predictable but $\langle S \rangle$ is (provided that we have determined the probabilities of the accessible states $\mathbf{A}_n$). Thus, it is of vital interest from the standpoint of model applications to know how far to *expect* S to fall from the ensemble mean value $\langle S \rangle$. Denoting the difference by

$$\varepsilon = S - \langle S \rangle \tag{20a}$$

we see that the ensemble mean square value of ε is

$$\langle \varepsilon^2 \rangle = \sum_{n=1}^{10{,}077{,}696} \text{Prob}\,(\mathbf{A}_n)(S_n - \langle S \rangle)^2. \tag{20b}$$

If $\langle \varepsilon^2 \rangle^{1/2}$ is a small fraction of $\langle S \rangle$, then within the ensemble the values of S are clustered around $\langle S \rangle$ and we can expect values near $\langle S \rangle$ to occur frequently. (The distribution of S_n shown in Fig. 5 is an example). This measure of the range of likely values is one of the easiest to determine and is used widely in statistical analyses.

In summary, we have shown that in systems possessing infinitely many degrees of freedom, such as the atmosphere, the combined information provided by physical laws and discrete space–time measurements of the state of the system on a given occasion are sufficient to delineate a *set* of *possible* states of the system (we called this the *scenario family* of states) but they are inadequate to specify which of these states the system is actually in at the time the measurements were made. The forces or mechanisms that determine the specific state are either unknown to us or they are beyond description. We proposed to model the combined action of all unspecifiable forces in a *specific* system by a *causal wheel* on whose circumference are inscribed numbers corresponding to the *accessible states*, which consist of some or all of the members of the scenario family of states. Before each realization of the system, we imagine that the wheel is spun by mother nature and the number on the wheel indicated by a fixed pointer when the wheel stops determines the actual state that the system acquires on that occasion. The angle of arc λ_n allocated on the wheel to state n, normalized by 2π, is defined as the *probability* of occurrence of state n. Probabilities can be determined

(approximately) only by observing the behavior of the specific system in question many times. From a large sequence of observations we learn how frequently we can *expect* a given state to occur. The concept of probability is an attempt to quantify the expected frequency of states, and with this knowledge we can refine our predictive capabilities somewhat by weighting the members of the scenario family of possible states by their probabilities. The average of a quantity over the scenario family in which the members have been weighted according to their probabilities is called an *ensemble mean* and is denoted by the angle brackets $\langle\ \rangle$. For example, the ensemble mean velocity $\mathbf{u}$ at a specific point $(\mathbf{x}_0, t_0)$ is defined as

$$\langle \mathbf{u}(\mathbf{x}_0, t_0) \rangle = \sum_i \text{prob}\,[\mathbf{u}(\mathbf{x}, t)_i]\,\mathbf{u}(x_0, t_0)_i, \quad (21)$$

where the summation is over *all* state trajectories $u(\mathbf{x}, t)_i$ passing through the points in $W^{MN}(t_0)$. Ensemble mean values like $\langle \mathbf{u} \rangle$ of state parameters are predictable, but the actual state $\mathbf{u}$ itself is *not* predictable. Therefore, the utility of a model in a particular problem depends on whether we can *expect* the actual state of the system to differ from the predictable mean state by an amount smaller than the error the model user is willing to allow. A measure of the expected errors is provided by the predictable quantity $\langle \varepsilon^2 \rangle$, defined by (20).

In Section 5 we will describe how the ensemble properties of concentration c are related to those of the fluid velocity $\mathbf{u}$.

4. FORMULATION OF AIR POLLUTION MODELS FOR APPLICATIONS

Equation (15) is the fundamental universal statement (or model) that we can make about material concentration in a fluid. This equation describes the effects on concentration of the natural laws of mass conservation and chemical transformation. Based on this equation we can make singular (deterministic) predictions of concentration values at any point in $(\mathbf{x}, t)$ provided that we can make singular statements about (1) the material concentration values everywhere in space at some time t_0; (2) the fluxes of material through all boundaries; (3) the rate $S_m(\mathbf{x}, t)$ at which each species $m = 1, \ldots, J$ is being injected into the fluid; (4) the transformation rate matrix k_{mij} and molecular diffusivities κ_m, $m = 1, \ldots, J$; and (5) the fluid velocity at all points $\mathbf{u}(\mathbf{x}, t)$.

In some instances we can fulfill the first four of these requirements. An example is the case of chemically inert materials ($k_{mij} \equiv 0$) with zero boundary fluxes, a clean atmosphere initially ($c_m(\mathbf{x}, t_0) = 0 \quad m = 1, \ldots, J$); and hypothetical sources S_m of each species. But as we have already discussed, we cannot make singular statements about the wind velocity $\mathbf{u}$. In fact, as it turns out, even with $c_m(\mathbf{x}, t_0)$, k_{mij} and κ_m, $S_m(\mathbf{x}, t)$ and the boundary fluxes prescribed, in practice we cannot make singular predictions of $c_m(\mathbf{x}, t)$ for individual members $\mathbf{u}(\mathbf{x}, t)$ of the flow ensemble. Because in the atmosphere the spatial and temporal variations in $\mathbf{u}$ span such vast ranges of scale (over 8 orders of magnitude in spatial scales), we cannot express the solutions $c_m(\mathbf{x}, t)$ of Equation (15) exactly. This problem is compounded when we consider photochemical materials, where $k_{mij} \neq 0$, because then the equation set has a higher degree of nonlinearity.

Thus, the essential task in formulating an air pollution model for practical applications is finding an analogue of the system of Equation (15) that is both solvable and universally valid. If these two requirements are mutually exclusive, i.e. if no solvable analogue exists that is universally valid, then air pollution modeling is not a viable approach to applied problems because the predictions of any given model could be in conflict with observations even if the singular statements used in the model were precise and the flow field $\mathbf{u}$ were known exactly.

The only solvable analogues of (15) are those that describe concentration 'systems' whose total number of degrees of freedom is compatible with the memory size of available computers and which require a computational effort that is compatible with the execution speed of available machines. We can generate a system with any desired degree of freedom from one with infinite freedom by performing suitable space–time averaging. And by applying the same averaging to the equations that describe the infinite system, we obtain the equations that govern the finite one. However, the system of equations that we produce by this process contains additional terms that involve the small scale details in the variables that we are trying to avoid. These terms represent additional unknowns in the equations and therefore they render the system undetermined. In order to 'close' the set of equations, we must invoke some assumption regarding the relationship of the unknown terms to the dependent variables in the equations and it is in the choice of this closure approximation that we are most likely to commit an error that will cause the resulting analogue of (15) to lack universal validity.

The best way to mitigate the impact of the closure approximation is to make the space–time averaging interval that is applied to Equation (15) as small as possible (see for example, Wyngaard, 1982). In this way essential features of important processes are treated explicitly in the equations rather than implicitly in the closure scheme. With current computers we can handle systems with the order of 10^6 degrees of freedom. Thus, in an application to photochemical air pollution where 20–40 chemical species had to be treated to resolve chemistry phenomena properly, one could allow 10^4–10^5 grid points to resolve variations in the concentration of each species in 3-D space. If this were a regional scale problem where the spatial domain of interest is of the order of $10^3 \times 10^3$ km in horizontal extent and 5 km deep, 10^5 grid points would provide a horizontal resolution of the order of 10 km and a

vertical resolution of 500 m, neither of which is adequate to resolve all the spatial variations that air pollutant concentrations are known to have. In this case the parameterizations of small scale processes that are adopted to achieve a model with only 10^6 degrees of freedom could be major sources of error. (The reader is referred to Lamb (1983) for a description of the techniques that were used to formulate the EPA's regional oxidant model.)

Our main point here is that the predictability of air pollutant concentrations is diminished by uncertainties arising from several sources. First, there is the uncertainty in the specification of the state of the atmosphere that we have discussed above. Second, there is the uncertainty introduced by the parameterizations that we use to reduce the concentration fields to analogues with finite degrees of freedom, i.e. the formulation of the model equations. Third, there is the error generated by the numerical technique used to solve the governing equations. And finally, there is the uncertainty arising from the errors in the measured parameter values, including the meteorological variables, that are inputs to the calculations.

In this paper we are concerned only with uncertainty arising from the first of these four sources, because of this group only the first is beyond our power to eradicate. We can improve our parameterization schemes, our numerical methods for solving differential equations, and the precision of our instruments, but we cannot acquire the infinite quantity of data that is necessary to specify the state of the atmosphere uniquely in any given region of space-time. Our aim is to formulate an approach to air pollution modeling that takes this inherent uncertainty into account in an explicit way. Toward this end we have shown that one can describe the velocity field of the atmosphere to within only a set of functions, defined by (14), that we have called metulence. For given emissions distributions $S_j(\mathbf{x}, t)$ of each of $j = 1, \ldots, J$ chemical species, the physical principles described by (15) that govern species concentrations in the atmosphere define a one-to-one mapping between each flow field in the metulence set and a concentration distribution $[c_j(\mathbf{x}, t)\ j = 1, \ldots, J]$. In the next section we show how the properties of the set C of concentration fields defined by this mapping are related to the set properties of the velocity fields from which C is produced.

5. RELATIONSHIP BETWEEN THE VELOCITY AND CONCENTRATION ENSEMBLES

We defined an ensemble earlier and pointed out that it is equivalent to the scenario family of states in which each member is weighted by its probability. The scenario family of concentrations c is obtained by using (15) to map each member of the scenario family of $\mathbf{u}$'s into the phase space of c. For brevity we will represent the metulence ensemble (of $\mathbf{u}$'s) by W and the corresponding ensemble of concentration by C, with the two related as shown in Fig. 4 with (15) used as the description of the laws of mass conservation and chemical transformation. An ensemble mean is therefore an average over the set W or C. (Note that in applications the phase space of C is finite-dimensional and the mapping between W and C is performed with the space–time averaged counterpart of (15) that we discussed earlier.)

We must examine the members of C to determine whether they possess some property, like the sum of the elements a_{ijn} of the matrices $\mathbf{A}_n$ that make up the ensemble of dice game outcomes, whose value can be expected to lie close to some predictable value, regardless of which member of C actually occurs. (In order to avoid awkward notation, we will use c in this Section to represent the members of C.)

Consider the time averaged value of each member $c(\mathbf{x}, t)$ of C at a given point $\mathbf{x}_0$, i.e.

$$\bar{c}(\mathbf{x}_0, t) = \frac{1}{2T}\int_{t-T}^{t+T} c(\mathbf{x}_0, t')\,dt', \quad c \in C \tag{22}$$

where $\mathbf{x}_0 \in D, T \leqslant t \leqslant \mathcal{T}-T$, and T is an arbitrary time interval.

Averaging (22) over the set C, we obtain the ensemble time averaged concentration

$$\langle \bar{c}(\mathbf{x}_0, t)\rangle = \frac{1}{2T}\int_{t-T}^{t+T} \langle c(\mathbf{x}_0, t')\rangle\,dt'. \tag{23}$$

Keep in mind that $\langle \bar{c}\rangle$ is the mean value of all possible time averaged concentrations $\bar{c}$ that could occur for a given source distribution $\mathbf{S}$ under the discrete set of observed meteorological variables that define the metulence (see Equation 14), represented by the ensemble W. Let

$$\varepsilon(\mathbf{x}_0, t) = \bar{c}(\mathbf{x}_0, t) - \langle \bar{c}(\mathbf{x}_0, t)\rangle \tag{24}$$

denote the difference between the time averaged value $\bar{c}$ of any one member of the ensemble and the (predictable) ensemble mean value $\langle \bar{c}\rangle$. Squaring (24) we get

$$\begin{aligned}\varepsilon^2(\mathbf{x}_0, t) = {} & \frac{1}{4T^2}\int\int_{t-T}^{t+T} c(\mathbf{x}_0, t')c(\mathbf{x}_0, t'')\,dt'\,dt'' \\ & - \frac{\langle \bar{c}(\mathbf{x}_0, t)\rangle}{T}\int_{t-T}^{t+T} c(\mathbf{x}_0, t')\,dt' \\ & + \langle \bar{c}(\mathbf{x}_0, t)\rangle^2.\end{aligned} \tag{25}$$

Averaging this expression over the ensemble C we obtain

$$\begin{aligned}\langle \varepsilon^2(\mathbf{x}_0, t)\rangle = {} & \frac{1}{4T^2}\int\int_{t-T}^{t+T} \langle c(\mathbf{x}_0, t')c(\mathbf{x}_0, t'')\rangle\,dt'\,dt'' \\ & - \langle \bar{c}(\mathbf{x}_0, t)\rangle^2.\end{aligned} \tag{26}$$

Note that $\langle \varepsilon\rangle = 0$.

The value of $\langle \varepsilon^2\rangle$ provides a measure of how closely the time averaged concentration $\bar{c}$ at $\mathbf{x}_0$ can be expected to lie to the predictable ensemble mean value $\langle \bar{c}\rangle$.

For example, from the Tchebycheff inequality of

statistics, we find that with $\langle \varepsilon \rangle = 0$ the fraction η of the ensemble population whose values $\bar{c}$ are within $\pm \varepsilon_0$ of $\langle \bar{c} \rangle$ is

$$\eta \geq 1 - \frac{\langle \varepsilon^2 \rangle}{\varepsilon_0^2}. \tag{27}$$

Thus, in a large number of predictions the average frequency with which the observed time average concentration $\bar{c}(\mathbf{x}_0, t)$ exceeds the predicted value $\langle \bar{c}(\mathbf{x}_0, t) \rangle$ by an amount larger than some fixed value ε_0 decreases as $\langle \varepsilon^2 \rangle$ decreases. Thus, our next task is to relate $\langle c(\mathbf{x}, t) \rangle$ and $\langle c(\mathbf{x}, t) c(x, t') \rangle$, the quantities needed to compute $\langle \varepsilon^2 \rangle$ and $\langle \bar{c} \rangle$, to the known ensemble properties of W.

In Lamb (1975) it is shown that for materials that are *chemically inert* or undergo at most *first order* reactions the ensemble mean concentration is given by

$$\langle c(\mathbf{x}, t) \rangle = \int\int_0 p(\mathbf{x}, t | \mathbf{x}', t') S(\mathbf{x}', t') \mathrm{d}t' \, \mathrm{d}\mathbf{x}' \tag{28}$$

and

$$\langle c(\mathbf{x}, t) c(\mathbf{x}, t') \rangle = \int\int\int\int_0^{t} p_2(\mathbf{x}, t; \mathbf{x}, t' | \mathbf{x}'', t''; \mathbf{x}''', t''') \times S(\mathbf{x}'', t'') S(\mathbf{x}''', t''') \mathrm{d}t'' \mathrm{d}t''' \mathrm{d}\mathbf{x}'' \mathrm{d}\mathbf{x}''', \tag{29}$$

where S is the source strength function of the material species whose concentration is c.

In (28) and (29) the functions p and p_2 are Lagrangian properties of the ensemble W, and hence it is these functions that link the properties of W and those of C for given S.

The function p is called the single particle displacement probability density and is defined for chemically inert material by

$$p(\mathbf{x}, t | \mathbf{x}', t') = \langle \phi(\mathbf{x}, t | \mathbf{x}', t') \rangle / \delta v. \tag{30}$$

Here, δv denotes the (small) sample volume centered at $\mathbf{x}$, $\langle\ \rangle$ denotes averaging over W, and ϕ is defined for each $\mathbf{u}(\mathbf{x}, t)$ in W by

$$\phi(\mathbf{x}, t | \mathbf{x}', t') = \begin{cases} 1, \text{ if } \mathbf{u}(\mathbf{x}, t) \text{ is such that a particle released at } (\mathbf{x}', t') \text{ is in } \delta v \text{ centered at } \mathbf{x} \text{ at time } t; \\ 0, \text{ otherwise.} \end{cases} \tag{31}$$

In the case of a point source of strength $S(t)$ located at $\mathbf{x}_0$, i.e.

$$S(\mathbf{x}, t) = S(t) \delta(\mathbf{x} - \mathbf{x}_0)$$

we obtain on combining (28) and (30)

$$\langle c(\mathbf{x}, t) \rangle = (\delta v)^{-1} \int_0 S(t') \langle \phi(\mathbf{x}, t | \mathbf{x}_0, t') \rangle \mathrm{d}t'. \tag{32}$$

The function p_2 that enters in (29) is called the two-particle displacement probability density and is defined as follows:

$$p_2(\mathbf{x}, t; \mathbf{x}, t' | \mathbf{x}'', t''; \mathbf{x}''', t''') = \langle \phi_2(\mathbf{x}, t; \mathbf{x}, t' | \mathbf{x}'', t''; \mathbf{x}''', t''') \rangle / (\delta v)^2. \tag{33a}$$

Here as in the definition of p (see 30) the angle brackets $\langle\ \rangle$ denote averaging over W; δv is the sample volume centered at $\mathbf{x}$; and ϕ_2 is defined for any $\mathbf{u}(\mathbf{x}, t)$ in W by

$$\phi_2(\mathbf{x}, t; \mathbf{x}, t' | \mathbf{x}'', t''; \mathbf{x}''', t''') = \begin{cases} 1, \text{ if } \mathbf{u}(\mathbf{x}, t) \text{ is such that a particle released at } (\mathbf{x}'', t'') \text{ and one released at } (\mathbf{x}''', t''') \text{ are found at } (\mathbf{x}, t) \text{ and } (\mathbf{x}, t'), \text{ respectively:} \\ 0, \text{ otherwise.} \end{cases} \tag{33b}$$

For a single point source at $\mathbf{x}_s$ the autocorrelation $\langle c(\mathbf{x}, t) c(\mathbf{x}, t') \rangle$ is derivable from the contracted form $\phi_2(\mathbf{x}, t;\ \mathbf{x}, t' | \mathbf{x}_s,\ t'';\ \mathbf{x}_s,\ t''')$. This function contains four independent variables for given $\mathbf{x}$ and $\mathbf{x}_s$ and is much more difficult to evaluate than ϕ.

Perhaps an example will help clarify some basic points. Suppose that we are interested in modeling a certain spatial region $\mathscr{D}$ during a specific time period $\mathscr{T}$. Suppose that at N arbitrary sites within $\mathscr{D}$ and at M time intervals in $\mathscr{T}$ we have error-free measurements of meteorological variables. Using special mathematical techniques, described qualitatively in Section 2 (see Lamb and Pelles, 1983 for details), we can generate from these data and physical laws jointly individual members of the set of metulence velocities $\mathbf{u}(\mathbf{x}, t)$, $\mathbf{x} \in \mathscr{D}$, $t \geq t_0$. And for each velocity we can calculate the trajectory that a fluid particle or pair of particles would follow released from $(\mathbf{x}_0, t_0)$.

As a specific example we consider the metulence set defined by (14) for $N = 80$ surface wind observations at a single hour ($M = 1$) on day 215 of 1979 within the region $\mathscr{D}$ considered in EPA's current regional modeling study. For simplicity, we shall assume that the flow field that exists at this hour is unchanging in time and that only the mass conservation equation (5) applies. Having generated the metulence set, say $\mathscr{W}$, defined by Equation (5) and these 80 wind observations, we find that $\mathscr{W}$ contains flow fields that are vastly unlike any flows that we have ever observed in the atmosphere. Some members of $\mathscr{W}$ have intense vortices between stations, some have strong jets that snake among the stations, and some exhibit other strange forms. Although these bizarre flows are *possible* under the conditions that we have specified, the fact that flows like them have never been observed in the atmosphere indicates that there are processes unaccounted for by Equation (5) and the 80 observations that effectively

preclude those flows from happening. The analogous situation with the dice game described earlier would be to find after many millions of games that certain members of the **A** family (illustrated in Fig. 5) never occur, perhaps because one of the dice we are using has the same number stamped on more than one of its faces. We have already pointed out that we can incorporate additional information that we have about the system by assigning a weight, called the probability, to each member of the scenario family, in this case the metulence set $\mathscr{W}$. To quantify our probability estimates, we note that it has been shown by both theory and measurements that the spatial variations of velocity in a viscous, 2-D fluid are of a scale such that the spectral distribution of kinetic energy is proportional to $|\mathbf{k}|^{-3}$, where $\mathbf{k}$ is the wave number of the fluctuation. Based on this empirical knowledge we propose that the probabilities of all members of $\mathscr{W}$ are zero except those flows in the subset W that lie along the intersections of the hypersurface $\mathscr{W}$ and the hypersurface, say Q, of functions whose Fourier transforms are consistent with the $|\mathbf{k}|^{-3}$ energy spectrum cited above.

Figures 7(a) and 7(b) show two members of the metulence subset W defined in this way. The flow field show in Fig. 7(a) is the field with minimum total kinetic energy from the infinite set W of flow fields $\mathbf{u}(\mathbf{x})$ each of which satisfies the 80 wind observations, is nondivergent and has a minus -3 energy spectrum.

A second member of the metulence set W is shown in Fig. 7(b). This velocity field, like the field shown in Fig. 7(a), satisfies all 80 observations exactly, it is horizontally nondivergent, and it has a minus -3 energy spectrum. If P_1 and P_2 designate (in velocity phase space) the flow fields shown in Figs 7(a) and (b), then as we have already noted, P_1 is the metulence field possessing minimum kinetic energy, i.e. the point in W nearest the origin; and P_2 is a point obtained by projecting an arbitrary vector, based at P_1, onto the hypersurface W. In both cases the simulated atmosphere has approximately 800 degrees of freedom in each velocity component: namely complex Fourier amplitudes for 40 wave numbers in x and 20 in y.

The particle trajectories indicated by the letter 'A' in Figs 7(a) and (b) originate from the site of one of the 80 meteorological stations, (indicated by the dark circles) and each is computed using the earlier assumption that the flow fields remain steady in time. Near the release point the two trajectories nearly overlap but with increasing distance downstream they diverge. At re-

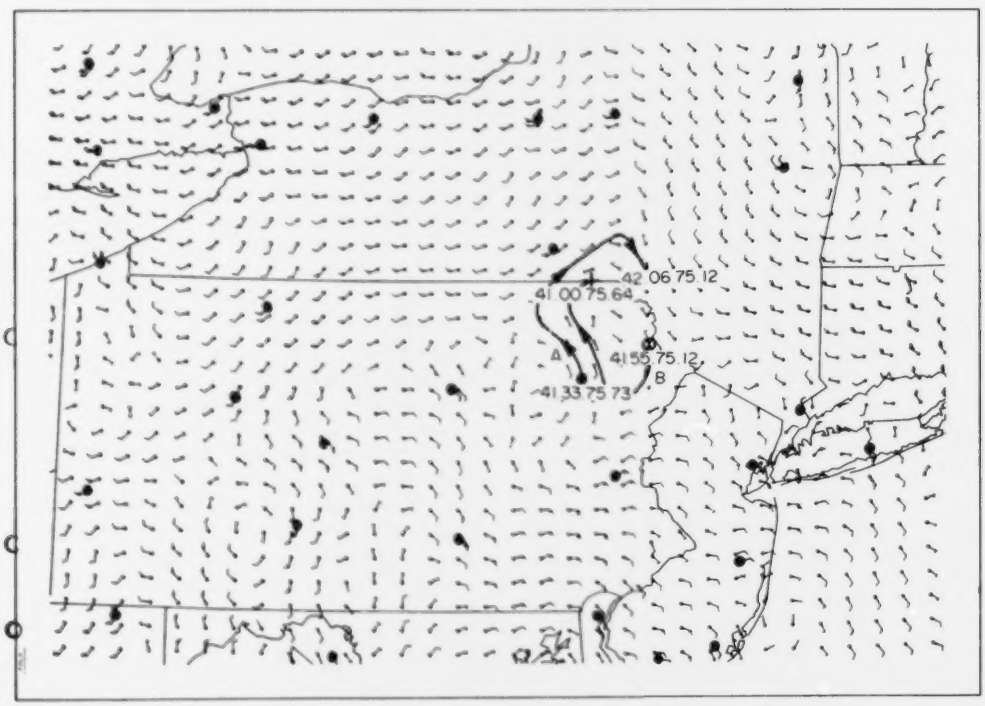

Fig. 7(a). Sample member of a metulence velocity set W based on 80 surface wind observations indicated by the heavy circles. (Most of the stations are outside the region shown.) The velocity field has approximately 800 degrees of freedom in each of its two components, it is horizontally non-divergent, and its energy spectrum satisfies a minus-3 power law. The trajectory labeled '*A*' originates at one of the sites of wind observations and assumes steady flow for a 15-h travel period. The trajectory labeled '*B*' is similar except it originates at a point of no wind data.

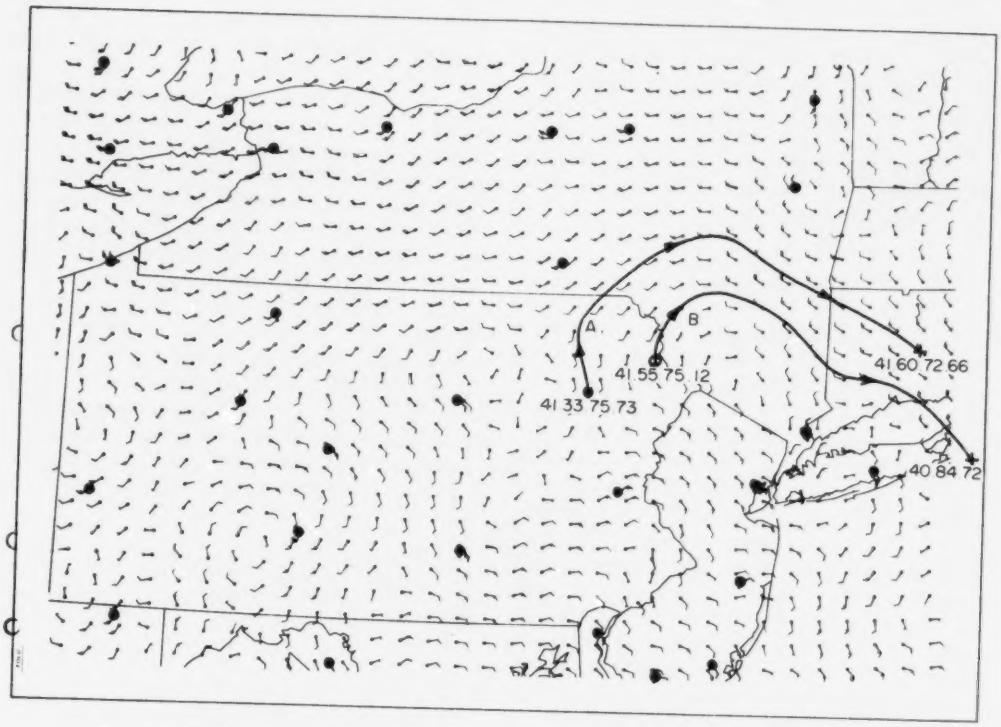

Fig. 7(b). Same as 7(a) except a different member of the same metulence set.

lease points other than one of the 80 measurement stations, the trajectories generally begin to diverge at the release point itself. The trajectories labeled 'B' in the figure are an example. This reflects the freedom of the flow fields in the scenario family at points not constrained by observations. All trajectories in Fig. 7 represent 15-h travel periods.

If two particles are released simultaneously from points close together, say from a 'point' source at $\mathbf{x}_0$, the rate at which they separate in time is determined by the energy in the small scale variations in the fluid velocity, specifically the scales comparable to the particle separation. In the atmosphere the small scale fluctuations in velocity are generally not large enough to cause a pair of particles to separate to a distance comparable to the separation of the 'A' trajectories shown in Figs 7(a) and (b). In other words, in the flow fields depicted in these figures, we would expect continuous plumes to remain rather slender, so that the plume corresponding to the 'A' trajectory in state 1 (Fig. 7a) would overlap that corresponding to state 2 (Fig. 7b) near the source, but beyond some travel distance the two plumes would become separated and would be widely separated at travel times corresponding to the end points of the 'A' trajectories shown in Figs 7(a) and (b).

Now if we were to plot the trajectories corresponding to other states of the metulence, for the same release point as the 'A' trajectories in Fig. 7, we would find that they all overlap near the release point (because the metulence is defined (see 14) in terms of a specific set of wind observations, one of which is taken at the point where the particles are released); but they diverge with increasing distance from this point. From this and the fact that pairs of particles released close together separate rather slowly, we can see intuitively that the square of the quantity ε, defined by 24, averaged over the concentration ensemble, i.e. $\langle \varepsilon^2 \rangle$ given by 26, will tend to be small within the immediate vicinity of a source situated near a meteorological station whose observations are used in the model. But $\langle \varepsilon^2 \rangle$ will increase with distance from the source, and it will be larger at all travel distances for sources that are not located near a meteorological station.

These observations suggest that the magnitude of the uncertainty in the concentration values predicted by a perfect model, arising from the uncertainty in the state of the atmosphere, is directly proportional to the extent to which concentration at the receptor is affected by distant sources. Furthermore, the level of uncertainty increases with distance to the nearest meteorological station, regardless of the proximity of the receptor to sources.

For example, in large urban areas where concentration levels are determined primarily by local sources, the concentrations c within the ensemble C will cluster around a single value, provided that some of the wind observations that define the metulence were made in

those areas. However, at points **x** far from sources, the values of $c(\mathbf{x})$ within the concentration ensemble will have a large scatter, regardless of the proximity of the receptor to a wind observation site, and hence one can expect observed concentrations in remote areas to differ significantly from the computed ensemble mean. An important task for future research is to quantify these uncertainty relationships.

6. CONCLUSIONS

We have shown that for a given source distribution **S** and a given set of meteorological observations, a model can delineate the *set* of functions C to which the actual concentration distribution $c(\mathbf{x}, t)$ resulting from those conditions belongs, but not the function $c(\mathbf{x}, t)$ itself. In dealing with second-order (nonlinear) reactants the only accurate way to determine the ensemble moments of the set C is to generate members of the set, by solving (15) for each of many members of the metulence ensemble W, and then to compute the set mean values $\langle c \rangle$, etc. by averaging the individual members. The conventional approach of formulating equations that yield ensemble moments directly encounters closure problems that are made virtually insurmountable by the combination of the nonlinearity of the chemistry and the large time and space scales of atmospheric motion.

From the ensemble C one can specify the frequency with which the actual concentration can be expected to fall within a given interval of values in a large number of observations. The width of this interval (see 27) is a measure of the 'predictability' of the concentration and it is determined by the properties of the set C. These in turn are determined by **S** and the set of meteorological data that define W. We speculated that predictability is greatest at receptors that are near both meteorological stations and sources. Predictability diminishes with increasing distance from *either* sources or meteorological stations. This means, for example, that there is inherently a limited ability to attribute pollutant levels in remote regions to particular sources far away.

Thus, the predictions even of a perfect model cannot be expected to agree with observations at all locations. Consequently, the common goal of 'model validation' should be one of determining whether observed concentrations fall within the interval indicated by the model with the frequency indicated, and if not, whether the failure is attributable to sampling fluctuations or is due to the failure of the hypotheses on which the model is based. From the standpoint of regulatory needs the utility of a model is measured partly by the width of the interval in which a majority of observations can be expected to fall. If the width of the interval is very large, the model may provide no more information than one could gather simply by guessing the expected concentration. In particular, when the width of the interval of probable concentration values exceeds the allowable error bounds on the model's predictions, the model is of no value in that particular application. An important task in this regard is the 'inverse' problem of specifying the density and type of meteorological data that are needed to achieve a given level of concentration predictability.

Acknowledgement—I am indebted to Dr. Don Pelles for generating the set of velocity fields illustrated in Fig. 7, and for his ongoing assistance in implementing the ideas presented in Section 2.

REFERENCES

Dold A. and Eckmann B. (editors) (1977) *Proc. Turbulence Seminar*, 1976/77. Vol. 615 of Lecture Notes in Mathematics, Springer, Berlin.

Lamb R. G. (1975) The calculation of long-term air pollutant concentration statistics using the concept of a macroturbulence. *Proc. IBM Seminar on Air Pollution Modeling* held in Venice, Italy, November 1975. IBM Venice Scientific Center, Report No. 48.

Lamb R. G. (1983) A regional scale (1000 km) model of photochemical air pollution—I. Theoretical formulation. EPA report EPA-600/3-83-035.

Lamb R. G. and Pelles D. (1984) The limits of determinism in atmospheric dispersion modeling (in preparation).

Papoulis A. (1965) *Probability, Random Variables and Stochastic Processes*. McGraw-Hill, New York.

Popper K. R. (1959) *The Logic of Scientific Discovery*. Basic Books, Inc., New York, NY.

Wyngaard J. C. (1982) Boundary layer modeling. In *Atmospheric Turbulence and Air Pollution Modeling* (edited by Nieuwstadt F. T. M. and van Dop H.) D. Riedel Publishing Co. Dordrecht, Holland, 69–106.

NOTICE

This document has been reviewed in accordance with U.S. Environmental Protection Agency policy and approved for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

This article documents research that was conducted under the auspices of the U.S. Environmental Protection Agency (USEPA). As an arm of the government, the USEPA has a paid up, non-exclusive, irrevocable, worldwide license to reproduce, prepare derivative works, and distribute to the public all of the material contained herein, and to grant others acting on its behalf the right to do so. The USEPA grants to the National Technical Information Service, Department of Commerce, the right to act as its agent in the distribution of this material.